

Growth and spin-resolved photoelectron spectroscopy of the MgO/Fe(110) system

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Abstract

Electronic and structural properties of ultrathin MgO layers grown on epitaxial Fe(110) films were investigated at room temperature by means of spin-resolved photoelectron spectroscopy, Auger-electron spectroscopy, and low energy electron diffraction. The spin polarization at the Fermi level of the Fe(110) film decreases abruptly with increasing thickness of MgO layer up to 7 Å. This behavior is attributed to the formation of a thin FeO layer at the MgO(111)/Fe(110) interface, attenuating the intrinsic spin polarization.

The high tunneling magnetoresistance (TMR) values achievable by means of magnetic tunnel junctions (MTJs) consisting of two ferromagnetic electrodes separated by a thin insulating layer have attracted strong interest for potential applications in magnetoelectronics [1–4]. According to Jullière’s model magnetoresistance of such MTJs depends only on the spin polarization of the ferromagnetic electrodes used. In contrast, *ab initio* electronic structure and transport calculations have shown that the magnetoelectronic properties of such devices strongly depend on the structural as well as electronic properties of the insulating layer and the specific termination at the insulator/ferromagnet (I/FM) interface [5–8].

In the last few years the epitaxial Fe/MgO/Fe(100) MTJ system has been intensively studied. For such ideal MTJs with abrupt interfaces between MgO and Fe, the TMR values are predicted to be as high as $\sim 2000\%$ [8]. Recent experiments on the Fe(100)/MgO/FeCo MTJ [9] and Fe/MgO/Fe MTJ [10] show a TMR value of only 60% at 30 K and 100% at 80 K, respectively, which are close to the theoretically predicted value of 75% for the system assuming a FeO layer at the MgO/Fe interface [11]. This fact can be considered as an indirect proof for the iron oxide formation at the inhomogeneous MgO/Fe interface. The surface x-ray diffraction (SXRD) and vibration spectroscopy experiments [12–14] which were carried out on the MgO/Fe(100) system gave a direct evidence for the formation of a FeO sub-monolayer at the interface.

In the present work the crystallographic and electronic structure of the MgO/Fe(110) interface were investigated by means of low energy electron diffraction (LEED), Auger electron spectroscopy (AES), and spin- and angle-resolved photoelectron spectroscopy (SPARPES). For this purpose MgO layers with (111) orientation can be epitaxially grown on the *bcc* *d*-metal film as it was shown earlier [15,16]. A TMR effect of about 30% with Fe(110) layers

as magnetic electrodes and amorphous Al_2O_3 layer as insulator was demonstrated by Yuasa *et al.* [17]. The present SPARPES experiments show that the spin polarization of photoelectrons at the Fermi level (E_F) decreased rapidly with increasing MgO layer thickness which can not only be due to the spin scattering of photoelectrons in the MgO layer. This fact can be attributed to the formation of a thin depolarizing FeO layer at the MgO/Fe interface on the basis of a spin-dependent transport model of iron valence band photoelectrons through the oxide overlayer.

All experiments were carried out at room temperature in a UHV system for energy-resolved SPARPES with spin analysis described in detail in Ref. [18]. It consists of a UHV chamber equipped with LEED optics, gas inlet, and an AES spectrometer with a cylindrical mirror analyzer. All AES spectra were recorded in the dN/dE mode with 2.5 keV primary electron energy and peak-to-peak modulation voltage of 2 V. The SPARPES spectra (He I, $h\nu=21.2\text{ eV}$) were recorded in normal emission by a 180° hemi-spherical energy analyzer connected to a 100 kV Mott detector for spin analysis. The energy resolution is 100 meV and the angle resolution $\pm 1^\circ$. The spin-resolved measurements have been performed in magnetic remanence after having applied a magnetic field pulse of about 500 Oe along the in-plane $\langle 1\bar{1}0 \rangle$ easy axis of the thin Fe(110) films [21].

Clean 50-Å thick Fe(110) films were prepared *in situ* by electron-beam evaporation onto a W(110) substrate, while the thickness was simultaneously monitored by a quartz microbalance. The degree of crystalline order of the thin epitaxial Fe(110) films and the MgO overlayer on top of Fe(110) films was checked by LEED. The surface cleanliness has been monitored by AES and valence band PES. The MgO was deposited *in situ* by electron beam evaporation from a W-crucible. The base pressure in the vacuum chamber was 1×10^{-10} mbar

and increased up to 1×10^{-9} mbar during the MgO deposition process.

Fig.1 shows LEED images of (a) the clean Fe(110) film as well as (b) the 30-Å thick MgO layer on top of the Fe(110) film. The well-ordered hexagonal (1×1) LEED structure of the MgO layer is clearly visible. For intermediate thicknesses of MgO (less than 30 Å) on top of the Fe(110) film the LEED images do not show any well-ordered structure, which means that a smooth transition from the *bcc* (1×1) structure to the *fcc* (1×1) structure takes place. The spots in these cases are very weak or not visible, with a diffuse background (not shown here).

The modification of the electronic structure of the MgO/Fe(110) system as function of the thickness of the deposited MgO layer was investigated by means of photoelectron spectroscopy (PES) in normal emission. The PES spectra obtained in this experiment are shown in Fig.2. They show a pronounced structure corresponding to the emission from the Fe ($3d$) states in the range of binding energies near E_F as well as from the O ($2p$) states in the range of 4-10 eV. With an increasing amount of deposited MgO a gradual shift of the maxima of the O ($2p$) states and the valence band edge (VBE) toward of larger binding energies can be clearly seen in Fig.2. In the inset of Fig.2 the change of the onset of the VBE position is shown with an increasing MgO layer thickness. This shift is approximately 1 eV. As it was discussed earlier by Kiguchi *et al.* [20] for the MgO/Ag(001) system this effect can be attributed to the increase of the binding energy of the Mg ($3s$) states above E_F and the increase of the binding energy of the O ($2p$) states, which results from the change in the Madelung potential for thin films.

The spin-resolved electronic structure of the MgO/Fe(110) system was studied by SPARPES. The spin-resolved photoemission spectra together with the total emission

intensity and the spin polarization as a function of the binding energy of Fe(110), 2 Å MgO/Fe(110) as well as 5 Å MgO/Fe(110) are presented in Fig.3a) (from bottom to top) and b), respectively. The spin-resolved spectra of the valence band of the Fe(110) film show the emission from the $\Sigma^1 \downarrow \oplus \Sigma^3 \downarrow$ states near 0.25 eV and from the $\Sigma^1 \uparrow \oplus \Sigma^4 \uparrow$ states near 0.7 eV. The value of the spin polarization of about $(-80 \pm 5\%)$ and the shape of the spectra are in agreement with previous measurements [21]. After the deposition of a 2-Å thick MgO layer on the Fe(110) film surface the total intensity of the photoemission spectra measured near E_F decreases drastically. At the same time, the features of the valence band of Fe can still be observed. For this system the spin polarization near E_F is decreased to about $-(52 \pm 5)\%$ compared to $-(80 \pm 5)\%$ of the clean Fe(110) surface. Additional deposition of MgO on top of the Fe(110) film leads to a further decrease of the spin polarization at E_F (shown for the 5 Å MgO/Fe(110) system in Fig.3 with $P(E_F) = -(21 \pm 5)\%$).

Fig.4 shows experimentally determined changes of the normalized spin polarization at E_F of the MgO/Fe(110) system as function of the deposited MgO layer thickness. In general, the changes of the spin polarization at E_F of the emitted photoelectrons in the system FM/oxide can be presented by the formula:

$$P = \frac{J_0^+ \exp(-\sigma^+ \cdot d) - J_0^- \exp(-\sigma^- \cdot d)}{J_0^+ \exp(-\sigma^+ \cdot d) + J_0^- \exp(-\sigma^- \cdot d)},$$

where $J_0^+(J_0^-)$ is the spin-up (down) photoelectron intensity without the oxide, d is the thickness of the oxide, and $\sigma^+(\sigma^-)$ is the scattering cross section for spin-up (down) electrons ($\sigma = \frac{\sigma^+ + \sigma^-}{2}$ is the averaged total scattering cross section [22]). Following this formula one can estimate the values for σ^+ and σ^- in the MgO/Fe(110) bilayer using the experimentally observed dependencies of P on $J^+(d)$, $J^-(d)$ and d yielding the values: $\sigma^+ = 1.5 \text{ nm}^{-1}$, $\sigma^- = 5.3 \text{ nm}^{-1}$ ($\sigma = 3.4 \text{ nm}^{-1}$). The average total scattering cross section value is clearly larger

than the value which can be expected for MgO. From the work of Siegmann [22] the value of about $\sigma=0.8 \text{ nm}^{-1}$ for MgO (only s and p electrons in the valence band) can be extracted. The scattering cross sections for spin-up and spin-down electrons in MgO are different (different \vec{k} values for spin-up and spin-down electrons in the valence band). On the basis of the observed significant difference of the estimated value of $\sigma=3.4 \text{ nm}^{-1}$ and the expected value of only $\sigma_{MgO}=0.8 \text{ nm}^{-1}$ one can suppose the formation of, e.g., a thin FeO layer at the MgO/Fe interface. The presence of such a FeO interfacial layer has been recently experimentally identified by AES and STM in MgO/Fe(100) and MgO/Fe(110) systems [23,24].

Following this assumption the spin polarization of emitted electrons can be written as

$$P = \frac{J_0^+ \exp(-\sigma_{FeO}^+ \cdot x) \exp(-\sigma_{MgO}^+ \cdot d) - J_0^- \exp(-\sigma_{FeO}^- \cdot x) \exp(-\sigma_{MgO}^- \cdot d)}{J_0^+ \exp(-\sigma_{FeO}^+ \cdot x) \exp(-\sigma_{MgO}^+ \cdot d) + J_0^- \exp(-\sigma_{FeO}^- \cdot x) \exp(-\sigma_{MgO}^- \cdot d)},$$

where σ_{FeO}^+ (σ_{FeO}^-) is the scattering cross section for spin-up (down) electrons in the thin FeO interface layer ($\sigma_{FeO} = \frac{\sigma_{FeO}^+ + \sigma_{FeO}^-}{2} = 3.5 \text{ nm}^{-1}$, four holes in the valence band of $\text{Fe}^{2+}\text{O}^{2-}$ [22]), σ_{MgO}^+ (σ_{MgO}^-) is the scattering cross section for spin-up (down) electrons in the MgO layer ($\sigma_{MgO} = \frac{\sigma_{MgO}^+ + \sigma_{MgO}^-}{2} = 0.8 \text{ nm}^{-1}$), and x is the thickness of the hypothetical FeO layer. From a comparison of the two equations for P the thickness of the FeO interfacial layer (x) can be estimated by:

$$x = d \cdot \frac{\sigma^+ + \sigma^- - 2\sigma_{MgO}}{2\sigma_{FeO}},$$

i.e. a linear dependence between the FeO interfacial layer thickness (x) and the MgO overlayer thickness (d).

The calculated upper limit values of the FeO interfacial layer thickness vs. MgO layer thickness are presented in the inset of Fig.4. This model is suitable in case of abrupt interfaces in the MgO/FeO/Fe(110) system and does not take into account scattering at the interfaces, at possible Fe inclusions in the MgO layer, or at defects etc. Thus the

FeO layer thickness can actually be smaller than estimated by our model. The formation and the increase of the FeO layer thickness at the MgO/Fe interface with increasing MgO layer thickness was also observed in recent AES experiments performed at the MgO/Fe(110) system [24].

In conclusion, the growth process of thin MgO films on Fe(110) and the electronic structure of the MgO/Fe(110) interface have been investigated. The SPARPES experiments show that the spin polarization of the Fe(110) thin film strongly decreases with an increasing MgO film thickness. This behavior can not be ascribed to the scattering process of the spin-polarized photoelectrons in only the MgO overlayer. In this case the formation of a thin iron oxide layer at the MgO/Fe interface was supposed which describes well the dependence of the spin polarization as function of the MgO overlayer thickness. An upper limit of the iron oxide layer thickness of two monolayers is deduced for an MgO layer of 7 Å.

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FIGURE AND TABLE CAPTIONS

Fig.1. LEED images of (a) a 50 Å thick Fe(110) film on a W(110) substrate and (b) 30 Å of MgO on top of Fe(110) film. The energy of the primary electron beam is 123 eV for (a) and 104 eV for (b). (the sixth reflex in both pictures is not visible due to the sample holder).

Fig.2. PES spectra ($h\nu=21.2$ eV) of the MgO/Fe(110) system as a function of the MgO layer thickness (shown on the right-hand side of the each spectra). The inset shows the change of the valence band edge (VBE) position of O ($2p$) states with increasing MgO thickness for the MgO/Fe(110) system.

Fig.3. (a) The spin-resolved photoemission spectra (spin down: down triangle, spin up: up triangle) together with the total emission intensity (circles) for Fe(110), 2 Å MgO/Fe(110), and 5 Å MgO/Fe(110) (from bottom to top). (b) The spin polarization as function of binding energy of a 50 Å thick Fe(110) film (solid square), 2 Å MgO/Fe(110) (open triangle up), and 5 Å MgO/Fe(110) (solid circle).

Fig.4. The change of the normalized spin polarization at E_F of the MgO/Fe(110) system with an increasing MgO layer thickness. The spline fit to the experimental data is shown by a dot-dashed line. The inset presents the estimated thickness of the iron oxide layer at the MgO-Fe interface as a function of MgO layer thickness.